VanVleck Response Of A Two-Level System And Mesoscopic Orbital Magnetism Of Small Metals

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We evaluate the mean value of the van Vleck response of a two-level system with level spacing distribution and argue that it describes the orbital magnetism of small conducting particles.

I. INTRODUCTION

The significance of the few-level physics for small metal particles has long been realized [1], [2]. At temperatures smaller than the mean level spacing at the Fermi level, $T \lesssim \Delta$, the spin susceptibility, for instance, is determined by the (spin-flip) transitions to the first unoccupied state for the particles that contain even number of electrons and is typically much smaller than the Pauli susceptibility. For the odd-electron particles, it is given by the Curie susceptibility. This is in contrast to the $T \gg \Delta$ (and the bulk) case where the leading term is given by the Pauli susceptibility [3]. Moreover, due to the exponential activation factors associated with transitions to unoccupied states, one should expect extremely broad distribution of susceptibility values and, therefore, large variations from particle to particle [4].

The orbital magnetic response of small metal particles, on the other hand, does not easily reduce to a few-level problem. While it does turn out to be essentially a Fermi level property for $T \gg \Delta$ [5] and in the bulk systems [6]-[8], a priori it appears as a property of the entire Fermi sea since all the levels are perturbed by the magnetic field. The two competing contributions to orbital magnetism are the precession diamagnetism and the polarization (van Vleck) paramagnetism [9]. While no rigorous description of the orbital magnetism exists for $T \lesssim \Delta$, it is nonetheless expected to be a Fermi level property as well due to the large Fermi sea cancellations between the diamagnetic and paramagnetic contributions. However, at the Fermi level the van Vleck part of the orbital response should be dominant and also the one that is very sensitive to variations of the energy level spacings. Consequently, we study a model wherein it is assumed that among all the "virtual transitions" between the ground and the excited states of the Fermi sea that are responsible for the van Vleck response, the one that determines the particle susceptibility is a single-electron "transition" from the last occupied to the first unoccupied state.

Imry has conjectured [10] that, since the electrons in a metal particle should be considered as a canonical ensemble [11], one can describe the particle response in terms of the difference between the magnetic energies of the canonical and grand canonical ensemble $F_H - \Omega_H$, the latter being the case for a bulk system and relatively well understood. In the limiting case of $T \lesssim \Delta$, one should expect the difference to be due to a single electron level crossing the position of the chemical potential in the equivalent grand canonical ensemble. However, the formalism based on this approach works only in the limit of $T \gg \Delta$, as explained in Refs. [3], [5], in which case $F - \Omega = \Delta \delta N^2/2$ where the particle number fluctuation in the equivalent grand canonical ensemble is actually much larger than one, $\delta N^2 \sim T/\Delta \gg 1$ [6]. In this limit, the perturbation theory can be used [12]. An improvement on this approach [5] involves using the exact, non-perturbative level correlation function and, in fact, yields a saturation value of the magnetic energy as T approaches Δ from above.

In what follows, we first give an argument in support of the large Fermi sea cancellation between the diamagnetic and paramagnetic contributions to the magnetic response. We then evaluate the two-level van Vleck response for the Fermi level and first unoccupied level. This involves averaging of the inverse energy spacing using the Gaussian Orthogonal Ensemble (GOE) statistics for energy eigenvalues and calculation of the magnetic dipole moment. The latter can be estimated using a semiclassical argument but also rigorously evaluated by considering the magnetic dipole absorption. We compare our result with the expression for the orbital magnetic response obtained in the limit $T > \Delta$. We restrict our analysis to 2D diffusive particles (disks, narrow strips, and rings) and use the units where $c = \hbar = 1$. We also neglect any sample-specific effects (fluctuations), except when specifically mentioned, and all the quantities in consideration are presumed disorder-averaged.

II. FERMI SEA CANCELLATION OF VAN VLECK PARAMAGNETISM AND PRECESSION DIAMAGNETISM

The van-Vleck energy of a two-level system in the magnetic field $\mathbf{H} = H_0\widehat{\mathbf{z}}$ is

$$\delta\epsilon_{vV}^{(1)} = \frac{\left|\langle i|\,\widehat{M}_z\,|f\rangle\right|^2 H_0^2}{\varepsilon_i - \varepsilon_f} \equiv \frac{\left|\widehat{M}_{if}\right|^2 H_0^2}{\varepsilon_i - \varepsilon_f} \tag{1}$$

and the total van Vleck energy is given by [9]

$$\epsilon_{vV}^{(tot)} = \frac{1}{2}\sum_{i,f}\left|\widehat{M}_{if}\right|^2 H_0^2 \frac{n_i - n_f}{\varepsilon_i - \varepsilon_f} \tag{2}$$

where $n_i = \theta(-\varepsilon_i)$. In the case of a continuous spectrum, we can rewrite eq. (2), using $dn_i/d\varepsilon_i = -\delta(\varepsilon_i)$, as

$$\epsilon_{vV}^{(tot)} = -\frac{1}{2}\upsilon H_0^2 \sum_f \left|\widehat{M}_{0f}\right|^2 = -\frac{1}{2}\upsilon\langle 0|\,M_z^2\,|0\rangle H_0^2 = -\frac{\upsilon e^2 v_F^2 \langle 0|\,r_\perp^2\,|0\rangle H_0^2}{16} \tag{3}$$

where $\upsilon \equiv \langle \upsilon(0)\rangle$ is the mean level density at the Fermi level, $\upsilon(\varepsilon)$ is the level density at energy ε. We have also used the fact that the magnetic moment at the Fermi level can be evaluated semiclassically, that is

$$\mathbf{M} = \frac{e}{2}\mathbf{r}\times\mathbf{v}_F \tag{4}$$

where $\mathbf{r}$ is the classical position vector of an electron and $\mathbf{v}_F$ is the Fermi velocity. In evaluating the coefficient in eq. (3), we averaged over the angle between $\mathbf{r}$ and $\mathbf{v}_F$.

The single-level diamagnetic energy diamagnetic energy is given by

$$\delta\epsilon_{diam}^{(1)} = \frac{e^2\langle i|\,r_\perp^2\,|i\rangle H_0^2}{8m} \tag{5}$$

and the total diamagnetic energy of the Fermi sea is given by [9]

$$\delta\epsilon_{diam}^{(tot)} = \sum_i \frac{e^2\langle i|\,r_\perp^2\,|i\rangle H_0^2}{8m} n_i \tag{6}$$

It is reasonable to conjecture that the disorder-averaged value $\langle i|\,r_\perp^2\,|i\rangle$ is i -independent which yields, upon converting to integration for a continuous spectrum,

$$\delta\epsilon_{diam}^{(tot)} = \frac{\upsilon e^2 v_F^2 \langle 0|\,r_\perp^2\,|0\rangle H_0^2}{16} \tag{7}$$

and is the same as eq. (3), with the opposite sign. This confirms the Fermi sea cancellation between the van Vleck paramagnetism and precession diamagnetism[1,2]. However, this derivation does not account for the quantum effects beyond the existence of the Fermi sea. Consequently, it is understood that the cancellation is not exact and that a Fermi level contribution is not accounted for in the present approximation. The nature of this contribution should depend on whether the chemical potential or the number of particles is fixed. In the former case, one expects a Landau response, as in bulk systems, while in the latter we make an ansatz of a two-level van Vleck response which involves the last occupied (Fermi) level and the first unoccupied level.

The situation is more complex in a strictly discrete level case where we will give only an order-of-magnitude argument. The first principles estimate of $\left|\widehat{M}_{if}\right|^2$ in the diffusive regime is based on the idea first proposed by Shapoval [13] and, later, applied by Gor'kov and Eliashberg [1] to small metal particles. Namely, we use the semiclassical approach to write

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[1]This argument can be carried over, with minor modifications, to 3D.

[2]For a disk of radius R, $\langle 0|\,r_\perp^2\,|0\rangle$ can be evaluated as the area average and equals to $R^2/2$.

$$\left|\widehat{M}_{if}\right|^2 = \frac{1}{\pi\upsilon}\overline{\int_0^\infty d\tau \exp\left[(\varepsilon_i - \varepsilon_f)\tau\right] M(t) M(t+\tau)} \tag{8}$$

where the bar denotes averaging over all classical trajectories. The correlation time scale τ for $M(t)$ is given by

$$\tau \sim \frac{\ell^2}{D} \tag{9}$$

where $\ell = v_F\tau$ is the electron mean-free-path and $D = v_F^2\,\tau/2$ is the diffusion coefficient. This is because the directions of $\mathbf{v}_F$ are uncorrelated after such time. Also, for $\varepsilon_f - \varepsilon_i > \tau^{-1}$ the exponential term becomes oscillatory. The scale of $\mathbf{r}$ is the relevant sample dimension a and we find,

$$\left|\widehat{M}_{if}\right|^2 \approx \frac{1}{\pi\upsilon}\overline{\int_0^\infty d\tau M(t) M(t+\tau)} \sim \frac{e^2\ell^2 v_F^2 a^2}{\upsilon D} \sim \frac{e^2 D a^2}{\upsilon} \sim \mu_B^2(\varepsilon_F\tau) \tag{10}$$

where μ_B is the Bohr magneton and ε_F is the Fermi energy. Consequently, the order of magnitude value of the two-level van Vleck response is obtained from eq. (1) as

$$\delta\epsilon_{vV}^{(1)} \sim -\frac{\left|\widehat{M}_{if}\right|^2 H_0^2}{\Delta} \sim -\frac{\mu_B^2 H_0^2}{\Delta}(\varepsilon_F\tau) \sim -|\chi_L| H_0^2 A(\varepsilon_F\tau) \tag{11}$$

where χ_L is the Landau susceptibility [6] and $A \sim a^2$ is the sample area. The total van Vleck energy can be estimated by multiplying $\delta\epsilon_{vV}^{(1)}$ by $(\tau\Delta)^{-1}$, which determines the range of the integral in eq. (8), and we find

$$\epsilon_{vV}^{(tot)} \sim -\frac{\mu_B^2 H_0^2}{\Delta}\frac{\varepsilon_F}{\Delta} \sim -|\chi_L| H_0^2 A\frac{\varepsilon_F}{\Delta} \tag{12}$$

Since $\langle 0| r_\perp^2 |0\rangle \sim a^2$, this is in qualitative agreement with eq. (3). Notice also that eq. (5) yields

$$\delta\epsilon_{diam}^{(1)} \sim \frac{\mu_B^2 H_0^2}{\Delta} \sim |\chi_L| H_0^2 A \tag{13}$$

for a single-level contribution to the precession diamagnetism.

III. VAN VLECK PARAMAGNETISM IN THE TWO-LEVEL MODEL

Turning again to eq. (1), where now ε_i is the energy of the last occupied state (Fermi level) and ε_f is the energy of the first unoccupied state, we find, averaging with the Wigner-Dyson distribution [14],

$$\delta\epsilon = -s\left|\widehat{M}_{if}\right|^2 H_0^2\frac{\pi}{2\Delta}\int_0^\infty \exp\left(-\frac{\pi x^2}{4}\right)dx = -\frac{\pi\upsilon}{2}\left|\widehat{M}_{if}\right|^2 H_0^2 \tag{14}$$

Here s is the level degeneracy ($s = 2$, on the account of spin) and $\upsilon = s\Delta^{-1}$. We have already estimated the matrix element using the semiclassical argument. However, a precise derivation of $\left|\widehat{M}_{if}\right|^2$ in the diffusive regime can be done by means of evaluation of the low-frequency magneto-dipole absorption in the field $\mathbf{H} = H_0\exp(-i\omega t)\,\widehat{\mathbf{z}}$. In the case of the continuous energy spectrum (for instance, when the level broadening γ is larger than Δ), the quantum-mechanical expression for the absorption should yield, up to small corrections, the classical value [15]. This is in complete analogy with the electric-dipole absorption (barring screening effects for the latter), which is discussed in detail in Ref. [16]. Since $\left|\widehat{M}_{if}\right|^2$ enters into the quantum-mechanical expression (see below), it can be extracted by equating with the classical value of the absorption.

The classical absorption is readily evaluated according to [17]

$$Q_{class} = \frac{1}{2}\omega\,\mathrm{Im}\left\{M_z^* H_0\right\} \tag{15}$$

where

$$M_z^{(disk)} = -\frac{AH_0}{4\pi}\left(1-\frac{2}{\varkappa R}\frac{J_1(\varkappa R)}{J_0(\varkappa R)}\right) \approx -\frac{AH_0}{32\pi}(\varkappa R)^2, \; A=\pi R^2 \tag{16}$$

$$M_z^{(strip)} = -\frac{AH_0}{8\pi}\left(1-\frac{\tan(\varkappa L_x/2)}{\varkappa L_x/2}\right) \approx -\frac{AH_0}{96\pi}(\varkappa L_x)^2, \; A=L_xL_y \tag{17}$$

for a disk of radius R and for a narrow metal strip, such that $L_x \ll L_y$, respectively. Here

$$\varkappa = \frac{1+i}{\delta}, \; \delta = \frac{1}{\sqrt{2\pi\sigma\omega}} \tag{18}$$

and σ is the Boltzmann conductivity. It is assumed that the frequency is such that $\delta \gg R, L_x$. Combining eqs. (15)-(18), we obtain

$$Q_{class}^{(disk)} = \frac{\omega^2 H_0^2 R^2 A\sigma}{16} \tag{19}$$

$$Q_{class}^{(strip)} = \frac{\omega^2 H_0^2 L_x^2 A\sigma}{48} \tag{20}$$

for the absorption, respectively, in the disk and the strip.

The quantum absorption, for the continuous spectrum, can be evaluated (in complete analogy with the electric-dipole absorption [16]) by means of the Fermi golden rule and we find

$$Q_{cont} = \frac{\pi}{2}\omega^2\upsilon^2 H_0^2\left|\widehat{M}_{if}\right|^2\frac{\langle\upsilon(0)\,\upsilon(\omega)\rangle}{\upsilon^2} \approx \frac{\pi}{2}\omega^2\upsilon^2 H_0^2\left|\widehat{M}_{if}\right|^2 \tag{21}$$

where the small quantum corrections of order Δ^2/γ^2 (or Δ^2/ω^2 if $\omega > \gamma$) [12] are neglected. For the electric-dipole absorption [1], [16], the quantity corresponding to $\widehat{M}_{if}$ is the electric dipole matrix element $\widehat{P}_{if}$. The latter can be evaluated from first principles in the diffusive approximation and the classical and quantum results can be evaluated independently and are equal in the considered limit. On physical grounds, we can assume that this is also true for the magnetic-dipole absorption. Consequently, we equate the r.h.s. of eqs. (19), (20) with that of eq. (21) and, with the use of $\sigma = e^2\upsilon D/A$, we obtain

$$\frac{\pi}{2}\upsilon\left|\widehat{M}_{if}^{(disk)}\right|^2 = \frac{e^2DR^2}{16} \tag{22}$$

$$\frac{\pi}{2}\upsilon\left|\widehat{M}_{if}^{(strip)}\right|^2 = \frac{e^2DL_x^2}{48} \tag{23}$$

for the disk and the strip respectively.

Substituting thus found value of $\left|\widehat{M}_{if}\right|^2$ in eq. (14), we find (for $s=2$) the following result for the van Vleck energy:

$$\delta\epsilon^{(disk)} = -\frac{1}{16}e^2DR^2H_0^2 \tag{24}$$

$$\delta\epsilon^{(strip)} = -\frac{1}{48}e^2DL_x^2H_0^2 \tag{25}$$

for the disk and the strip respectively. Eqs. (24) and (25) should be compared with the magnetic part of the energy obtained for $T > \Delta$ in the so called "mixed approximation" [5] wherein the exact, non-perturbative level correlation function is used yet the large particle-number fluctuation in the equivalent grand canonical ensemble is assumed also, the latter being true only for $T \gg \Delta$,

$$\delta\epsilon_{>} = -\frac{1}{2\pi}\tau_H^{-1} \tag{26}$$

The procedure for the evaluation of τ_H^{-1} using the gauge where the vector potential is tangential to the surface is described in Ref. [18] and we find

$$\delta\epsilon_{>}^{(disk)} = -\frac{1}{4\pi}e^2DR^2H_0^2 \tag{27}$$

$$\delta\epsilon_{>}^{(strip)} = -\frac{1}{6\pi}e^2DL_x^2H_0^2 \tag{28}$$

for the disk and the strip respectively.

We also mention the Aharonov-Bohm response of a narrow ring threaded by the flux ϕ, where the derivation is particularly simple. Namely, the van Vleck energy in this case is given by

$$\delta\epsilon_{if} = -\frac{|\widehat{v}_{if}|^2}{\varepsilon_i - \varepsilon_f}\left(\frac{e\phi}{2\pi R}\right)^2 \tag{29}$$

where $|\widehat{v}_{if}|^2$ is evaluated by considering absorption in the alternating flux $\phi\exp(-i\omega t)$ which generates the electric field according to the Lenz's law that, in turn, produces the Boltzmann current density proportional to v. Consequently, we find

$$|\widehat{v}_{if}|^2 = \frac{D}{4\pi\upsilon} \tag{30}$$

and, averaging over the level spacing,

$$\delta\epsilon^{(ring)} \sim -\frac{D}{8}\left(\frac{e\phi}{\pi R}\right)^2 \tag{31}$$

The latter has the same parametric dependence as

$$\delta\epsilon_{>}^{(ring)} = -\frac{1}{2\pi}\tau_H^{-1} = -\frac{D}{2\pi}\left(\frac{e\phi}{\pi R}\right)^2 \tag{32}$$

which is the limiting value of energy for $T > \Delta$ [5].

IV. DISCUSSION

The main result of this paper is that the parametric dependence of eqs. (24), (25) and (27), (28) is the same (with closely matching numerical coefficients). Consequently, the orbital magnetic response of a level-quantized metal particle can be satisfactorily explained in terms of the two-level van Vleck response. This conclusion also holds for other level distributions characteristic of disordered (chaotic) systems, such as the Gaussian Unitary Ensemble (only the numerical coefficient will be different).

Whereas the two-level picture might be also valid for the Poisson distribution, which is the case for classically integrable systems, the mean response needs to be examined more carefully because level "bunching," characteristic of this distribution, implies that for any magnitude of the field there will be a significant number of particles for which the perturbation theory no longer applies.

Another question which we hope to address in a future work is the sample-specific (fluctuation) effects that are expected to be quite large in the limit considered here. We point out that, in addition to the fluctuation of the level spacing and the magnetic moment, the details of the cancellation between the van Vleck paramagnetism and Landau diamagnetism should differ from particle to particle leading to variations in the number of levels contributing to the total response.

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